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Investigation of Hyperfine Interactions in GdNiIn Compound

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Abstract. Perturbed gamma–gamma angular correlation technique was used to measure the hyperfine interactions in the compound GdNiIn using the ^{111}In→^{111}Cd and ^{140}La→^{140}Ce probe nuclei at the In and Gd sites, respectively. A unique quadrupole frequency with asymmetry parameter η = 0.78 was observed for ^{111}Cd probe at In sites for the measurements above Curie temperature. Below T_C, the spectra for ^{111}Cd show combined magnetic dipole and electric quadrupole interaction. Below 85 K, a unique magnetic interaction is observed at ^{140}Ce. A linear relationship between the saturated magnetic hyperfine field and the magnetic transition temperature was observed for both probes, indicating that the main contribution to the mhf comes from the conduction electron polarization.

Key Words: magnetic hyperfine field, PAC spectroscopy, quadrupole interaction, rare earth magnetism.

1. Introduction

An important group within the series RTX (R = rare earth metal, T = transition metal, X = sp-element) is formed by compounds, which crystallize in the ZrNiAl-prototype structure (hexagonal structure for space group $P\bar{6}2m$) and show interesting magnetic properties and a variety of magnetic structures [1, 2]. This structure is formed by magnetic RT layers alternated with non-magnetic TX layers. The magnetic R atoms occupy the positions: x, 0, 1/2; 0, x, 1/2; $\bar{x}$, $\bar{x}$, 1/2 and form a triangular structure, which is a deformed Kagomé lattice. One of the characteristics of such a triangular arrangement of magnetic atoms is the frustration of the magnetic interactions when an antiferromagnetic order is present. The RNiIn family of compounds, which also crystallize in the ZrNiAl-type structure, has been very little studied so far. In this family, GdNiIn orders ferromagnetically below around 94 K as reported by Merlo *et al.* [3] using magnetization measurements. Canepa *et al.* [4] in the measurements of the Magnetocaloric properties observed a ferromagnetic transition at 96 K. Tyvanchuk *et al.* in AC

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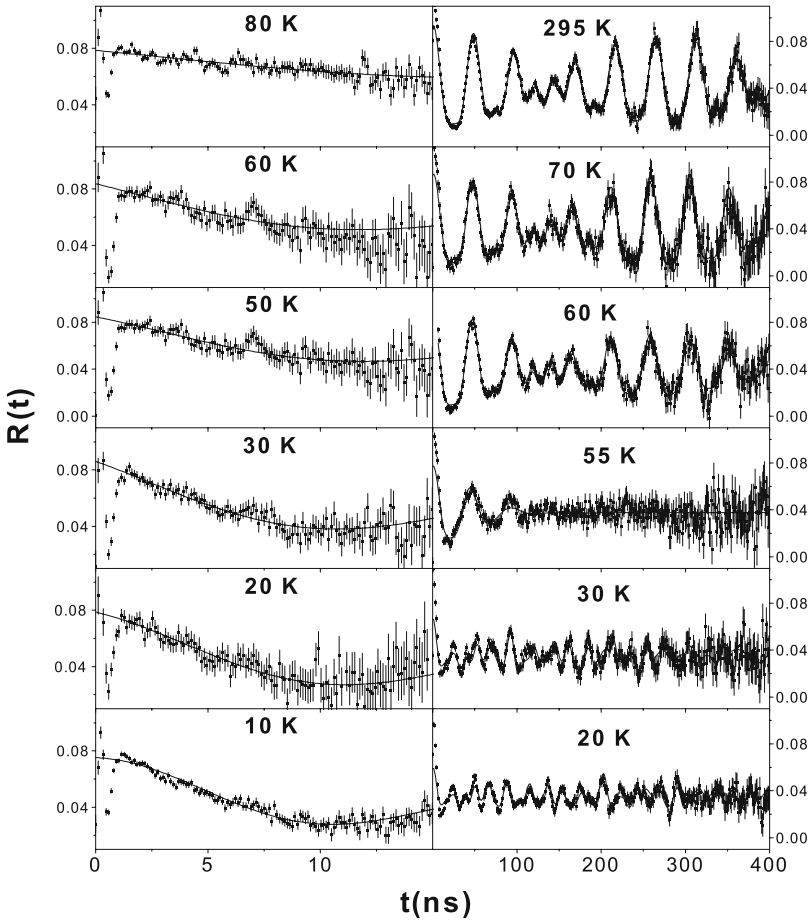

Figure 1. TDPAC spectra of the hyperfine interactions at Gd and In sites with ^{140}Ce (left) and ^{111}Cd (right) probes, respectively.

and DC bulk magnetic measurements in the compound GdNiIn reported an anomalous behavior at 80 K [5]. In this reference it was reported that because RNiIn compounds are good electrical conductor and there is a large interatomic distance between *R* atoms, the observed magnetic ordering would be caused by interactions via conduction electrons described by the RKKY model. In the present work, we have investigated the temperature dependence of the magnetic hyperfine field (mhf) on both Gd and In sites using ^{140}Ce and ^{111}Cd probes, respectively, as well as the behavior of the electric field gradient efg at In sites.

2. Experimental

The polycrystalline GdNiIn samples were prepared by repeatedly melting the constituent elements (Gd 99.99%, Ni 99.998%, In 99.9999%) in an arc furnace

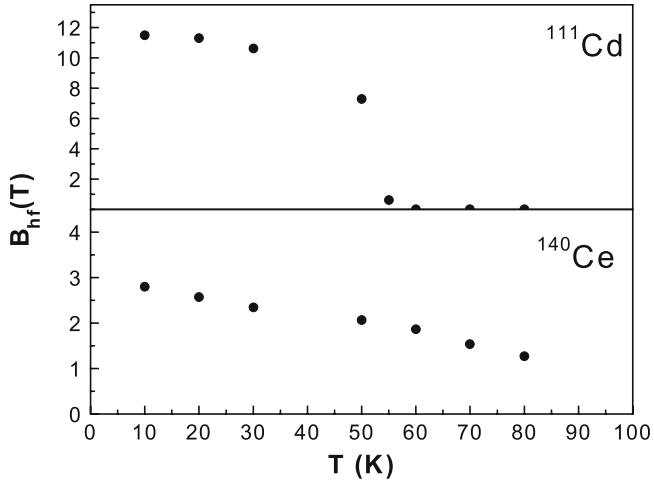

Figure 2. Temperature dependence of the magnetic hyperfine field at In (top) and Gd (bottom) sites in GdNiIn with ^{111}Cd and ^{140}Ce probes, respectively.

under argon atmosphere purified with a hot titanium getterer. Carrier free ^{111}In nuclei were introduced into the sample by diffusion. Another sample was prepared in a similar way but with radioactive ^{140}La nuclei obtained by neutron irradiation of lanthanum metal substituting about 0.1% of Gd atoms melted along with the constituent elements. Samples were annealed in vacuum for 72 h at 800°C. The structure of the samples were checked by X-ray diffraction measurement, which indicated a single phase and ZrNiAl-type structure with the $P\bar{6}2m$ space group for the compound.

The TDPAC measurements were carried out with a conventional fast–slow coincidence set-up with four conical BaF2 detectors. The well known gamma cascade of 172−245 keV, populated from the decay of ^{111}In with an intermediate level with spin $I = 5/2^+$ at 245 keV ($T_{1/2}$ = 84.5 ns) in ^{111}Cd, was used to investigate the hyperfine interactions in GdNiIn samples. The gamma cascade of 329−487 keV populated from the decay of ^{140}La with an intermediate level with spin $I = 4^+$ at 2083 keV ($T_{1/2}$ = 3.45 ns) in ^{140}Ce was used to measure the magnetic hyperfine field (B_{hf}) at Ce. The samples were measured in the temperature range of 10–420 K by using a closed-cycle helium cryogenic device. The time resolution of the system was about 0.6 ns for both gamma cascades. A detailed description of the method can be found elsewhere [6, 7].

3. Results and discussion

Some of the TDPAC spectra measured with ^{140}Ce and ^{111}Cd probe nuclei are shown in Figure 1. The solid curves are the least squares fit of the experimental data to the appropriate function in each case. The quadrupole moment of the

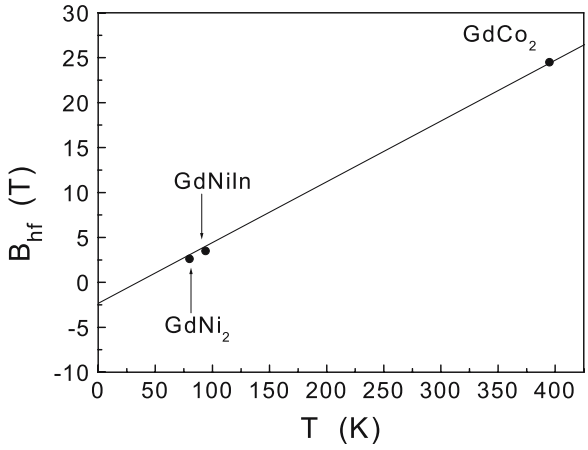

Figure 3. The extrapolated magnetic hyperfine field $B_{hf}(0)$ at ^{140}Ce in some Gd compounds as a function of the respective magnetic transition temperatures.

2083 keV 4^+ state of ^{140}Ce is known to be very small [8], consequently one expects to observe an almost pure magnetic dipole interaction in the antiferromagnetic phase of the sample. Below approximately 85 K, a unique magnetic interaction is observed at ^{140}Ce at Gd sites of GdNiIn. The temperature dependence of B_{hf} for ^{140}Ce is shown in the lower part of Figure 2. One can observe that there is a slight increase in the B_{hf} values at low temperature where the hyperfine field should be saturated. It is not clear if this increase is real because the B_{hf} values are quite low and, consequently, less precise.

The TDPAC spectra for ^{111}Cd at room temperature shows a unique quadrupole interaction with a sharp frequency ν_Q = 82.9(3) MHz, δ = 1% and η = 0.78(5). Below 60 K, the spectra for ^{111}Cd show combined magnetic dipole and electric quadrupole interaction. These spectra are characterized each by a single quadrupole frequency (ν_Q ~84 MHz) with δ = 1% and η = 0.78, and a temperature dependent magnetic dipole interaction shown in Figure 2. The angle between the efg and mhf changes from 60° at 50 K to 30° at 20 K. A comparison of the temperature dependence of the B_{hf} for both probes shows a significant difference in the transition temperature, around 55 K and ~85 K for ^{111}Cd and ^{140}Ce, respectively. We have no explanation for this difference yet. The values of the mhf at low temperature for ^{111}Cd are four times greater than the values for ^{140}Ce. This fact is mainly due to a difference in the distance of the probe to the magnetic Gd ion (3.2 Å for ^{111}Cd-Gd and 3.9 Å for ^{140}Ce-Gd).

In [9], the saturation values of B_{hf} at ^{111}Cd on Gd sites in $GdNi_2$, $GdAl_2$ and Gd are compared to the respective Curie temperature and the result showed a linear dependence where B_{hf}/T_C = 0.116 T/K. Using 11.5 T as the saturation value for B_{hf} in GdNiIn, the ratio B_{hf}/TC ~0.12 T/K and follows the same linear behavior reported in [9]. The saturation values of B_{hf} on Gd sites in GdNiIn, $GdNi_2$ (Carbonari A. W., private communication), and $GdCo_2$ [10] are compared

to the respective transition temperature of each host in Figure 3. One can observe that to a good approximation B_{hf} is a linear function of the transition temperature. According to the RKKY theory of indirect coupling the ratio between the conduction electron spin polarization (CEP) and the order temperature is expected to be proportional to $[J_{sf}(g-1)(J+1)]^{-1}$, where J_{sf} is the s-f coupling constant, g the Landé factor and J the total angular momentum. The linear relation between B_{hf} at ^{140}Ce and the magnetic transition temperature in Figure 3 thus may imply that the main contribution to the B_{hf} comes from the CEP at the probe site and the coupling constant J_{sf} has the same value in GdNiIn, $GdNi_2$ and $GdCo_2$ compounds. Therefore, the ^{140}Ce probes in this case behaves as closed shell nuclei like ^{111}Cd. Preliminary *ab initio* calculations using the WIEN2K code have shown that the main contribution to the mhf in GdNiIn using ^{111}Cd as impurity at In sites comes from valence electrons.

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